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Theory of Electric Polarization in Multiorbital Mott Insulators

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The interaction between the electric field **E** and spins in multiorbital Mott insulators is studied theoretically. We find a generic coupling mechanism, which works for all crystal lattices and which does not involve relativistic effects. It couples **E** to the "internal" electric field **e** originating from the dynamical Berry phase. We discuss several effects of this interaction: (i) an unusual electron spin resonance, (ii) the displacement of spin textures in an applied electric field, and (iii) the resonant absorption of circularly polarized light by Skyrmions, magnetic bubbles, and magnetic vortices.

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Introduction.—The manipulation of magnetic patterns with an applied electric field is an important issue both for fundamental physics and for applications to spintronics [1]. Naively, electrons in Mott insulators should be inert to the electric field oscillations with energies below the charge gap. Yet, a number of multiferroic materials, in which electric polarization is induced by spin orders [2], show strong response to the electric field at frequencies of magnetic excitations, resulting in the so-called electromagnon peaks in optical absorption [3,4].

Two main mechanisms for the coupling between the electric field and spins have been identified so far [5–8]. One is the lattice or electronic polarization induced by the Heisenberg spin exchange energy, which gives rise to the "bond" electric dipoles $\mathbf{P}_{ij}$ proportional to the scalar products of spins: $P^a_{ij} = \pi^a_{ij}\mathbf{S}_i \cdot \mathbf{S}_j$ [8]. The other originates from the relativistic spin-orbit interaction inducing the dipole moments proportional to the vector products of spins, $\mathbf{P}_{ij} = \alpha \mathbf{e}_{ij} \times (\mathbf{S}_i \times \mathbf{S}_j)$, where $\mathbf{e}_{ij}$ is the unit vector parallel to the bond [5–7]. The effectiveness of these mechanisms is restricted by symmetry requirements to special lattice geometries and magnetic orders, such as cyloidal spirals and the antiferromagnetic E-type order in orthorombically distorted manganites. One of the motivations for this study is to find a generic mechanism that couples electric field to spin patterns in insulating ferromagnets independently of their crystal structure.

Another motivation is the recent upsurge of interest in Mott insulators close to the transition into metallic state, e.g., $3d$ and $5d$ transition metal oxides [9,10], organic crystals [11], cold atoms [12], and quantum dot arrays [13]. The proximity to metallic state enhances fluctuations of the electron charge density playing the crucial role in the coupling of the low-energy spin degrees of freedom to the electric field. Furthermore, as we show below, large distances between the localized charges in artificial Mott insulators [13] can strongly amplify their response to an applied electric field.

In this Letter, we study theoretically the electric polarization induced by time-dependent spin patterns in magnetic insulators taking into account the spin dynamics during the exchange process. We derive the magnetoelectric coupling for a multiorbital Hubbard model and show that the most universal mechanism that does not require special crystal lattices and relativistic effects gives the electric polarization proportional to the "internal" electric field $\mathbf{e}(\mathbf{x}, t)$ associated with the Berry phase of dynamical spins,

$$\mathbf{e}\,(\mathbf{x}, t) = (1/2)\sin\theta(\partial_t\theta\boldsymbol{\nabla}\varphi - \partial_t\varphi\boldsymbol{\nabla}\theta). \tag{1}$$

This quantity has been discussed in the context of the "electromotive force" or "spin motive force" in metallic ferromagnetic systems [14–16]. We show that this field is also relevant to the insulating magnets contributing to their dielectric response. In particular, the coupling between external and internal electric fields makes it possible to shift noncollinear spin textures in ferromagnets by applying voltage.

Model.—Our microscopic model includes the sum of electron Hamiltonians on transition metal sites,

$$H_{\text{site}} = U\frac{n(n-1)}{2} + \Delta n_\beta - J_H[\mathbf{S}\cdot(\mathbf{s}_\alpha + \mathbf{s}_\beta) + \mathbf{s}_\alpha \cdot \mathbf{s}_\beta], \tag{2}$$

where the first term is the on-site Coulomb repulsion, $n = n_\alpha + n_\beta$ being the total number of itinerant electrons on the site, $\Delta > 0$ is energy splitting between the two orbitals α and β, and the last term is Hund's rule coupling between the local spin $\mathbf{S}_i$ and the spin $\mathbf{s}_{ia} = \frac{1}{2}c^\dagger_{ia}\boldsymbol{\sigma}c_{ia}$ of the itinerant electron on the orbital $a = \alpha, \beta$. The zeroth-order Hamiltonian is the sum of all on-site energies and H_S describing spin interactions that do not originate from exchange processes.

We do perturbation theory in the hopping energy of itinerant electrons,

$$V = -\sum_{ia;jb} t_{jb,ia} c^{\dagger}_{jb} c_{ia}, \tag{3}$$

where $t_{jb,ia} = t^{*}_{ia,jb}$ is the amplitude of hopping from the orbital a on the site i to the orbital b on the site j. In the presence of electric field the hopping amplitudes are modified using the "Peierls substitution" [17],

$$t_{jb,ia} \rightarrow t_{jb,ia} e^{-ie\int_{\mathbf{x}_i}^{\mathbf{x}_j} d\mathbf{x}\cdot\mathbf{A}(\mathbf{x},t)}, \tag{4}$$

where $\mathbf{A}$ is the vector potential, $-e$ is the electron charge, and $\hbar = c = 1$ (furthermore, the term $-eA_0 n$ is added to the on-site Hamiltonian).

In general, the electric field dependence of hopping amplitudes is more complex. It can originate from a nonzero electric dipole moment of the metal-ligand-metal bond, which gives rise to the exchange striction and relativistic mechanisms discussed in the Introduction [5–8]. Here we only take into account the E dependence resulting from the Peierls substitution present for any bond geometry.

The model with two orbitals per site is the simplest model of a multiorbital Mott insulator. We first show that the electron hopping to an unoccupied orbital on a neighboring site (see Fig. 1), favoring ferromagnetic interactions between spins, gives rise to a dynamical electric polarization induced by rotating spins. Then we explain why this does not happen in the single-orbital Hubbard model. For simplicity we assume that there is only one electron per site and consider the strong Hund's rule coupling and large S limit, in which the electron spin is parallel to the local spin.

Electric polarization of spins.—Consider first the ferromagnetic model, in which only $t_{2\beta,1\alpha} = t_{1\beta,2\alpha} = t \neq 0$ (see Fig. 1). To second order in the hopping amplitude t, the correction to the imaginary time spin action is

$$\delta S = t^2 \int_0^{\beta} d\tau_i \int_{\tau_i}^{\beta} d\tau_f e^{-U'(\tau_f-\tau_i)} [C_{21}c_{21} + C_{12}c_{12}], \tag{5}$$

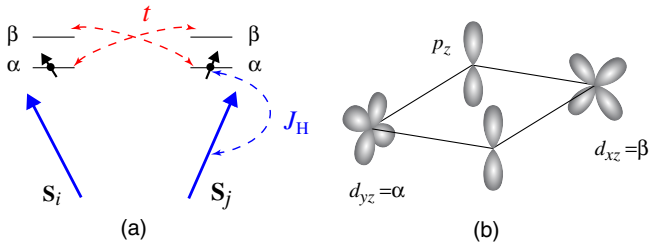

FIG. 1 (color online). (a) Two-orbital model of electrons interacting with the local spins $\mathbf{S}_i$ and $\mathbf{S}_j$ through the Hund's rule coupling J_H. (b) Physical realization of the two-orbital model describing the hopping between the $\alpha = d_{yz}$ and $\beta = d_{xz}$ orbitals of the magnetic ions on the metal-oxygen plaquette mediated by the p_z orbitals of oxygen ions.

where $U' = U + \Delta - J_H/4$,

$$C_{21} = e^{-ie\int_{\tau_i}^{\tau_f} d\tau \int_{\mathbf{x}_1}^{\mathbf{x}_2} d\mathbf{x}\cdot\mathbf{E}},$$
$$c_{21} = e^{\int_{\tau_i}^{\tau_f} d\tau [\langle \mathbf{n}_1(\tau)|\partial_\tau|\mathbf{n}_1(\tau)\rangle - \langle \mathbf{n}_2(\tau)|\partial_\tau|\mathbf{n}_2(\tau)\rangle - \delta H_{21}]} \times \langle \mathbf{n}_1(\tau_f)|\mathbf{n}_2(\tau_f)\rangle \langle \mathbf{n}_2(\tau_i)|\mathbf{n}_1(\tau_i)\rangle, \tag{6}$$

with $\delta H_{21} = \frac{1}{2(S+1/2)}(\mathbf{n}_2 \cdot \frac{\partial}{\partial \mathbf{n}_2} - \mathbf{n}_1 \cdot \frac{\partial}{\partial \mathbf{n}_1}) H_S$ describing the spin energy change due the electron hopping from site 1 to site 2 (C_{12}, c_{12} are obtained by interchanging the indices 1 and 2). Here, $\mathbf{n}_{1,2}$ is the unit vector in the direction of the local spin $\mathbf{S}_{1,2}$ and $|\mathbf{n}\rangle$ denotes the state of the electron with spin parallel to $\mathbf{n}$.

C_{21} is invariant under local gauge transformations and so is c_{21}. Introducing the vector potential of the "internal" field by $a_0 = i\langle \mathbf{n}|\partial_\tau|\mathbf{n}\rangle$ and $\mathbf{a} = i\langle \mathbf{n}|\partial_{\mathbf{x}}|\mathbf{n}\rangle$, we can write the overlap of the electron spin wave functions in the form

$$\langle \mathbf{n}_2|\mathbf{n}_1\rangle = \cos\frac{\theta_{21}}{2} e^{i\int_{\mathbf{x}_1}^{\mathbf{x}_2} d\mathbf{x}\cdot\mathbf{a}}, \tag{7}$$

where θ_{21} is the angle between $\mathbf{n}_1$ and $\mathbf{n}_2$. The vector $\mathbf{n}(\mathbf{x})$ that defines $\mathbf{a}$ varies between $\mathbf{n}_1$ to $\mathbf{n}_2$ along the shortest arc on the unit sphere, while $\mathbf{x}$ varies from $\mathbf{x}_1$ to $\mathbf{x}_2$. We now can write c_{21} in the manifestly covariant form

$$c_{21} = \cos\frac{\theta_{21}(\tau_f)}{2} \cos\frac{\theta_{21}(\tau_i)}{2} e^{\int_{\tau_i}^{\tau_f} d\tau (i\int_{\mathbf{x}_1}^{\mathbf{x}_2} d\mathbf{x}\cdot\mathbf{e} - \delta H_{12})}, \tag{8}$$

where $\mathbf{e} = \partial_{\mathbf{x}} a_0 - \partial_\tau \mathbf{a}$ is the gauge invariant internal electric field. Comparing Eqs. (6) and (8), we find that the correction to action only depends on the combination of applied and internal electric fields, $e\mathbf{E} - \mathbf{e}$.

Since the time spent by the hopping electron on a neighboring site, $\tau_f - \tau_i \sim (U')^{-1}$, is much shorter than the characteristic time of spin dynamics, C_{ij} and c_{ij} in Eq. (5) can be expanded in powers of $\tau_f - \tau_i$, which generates an expansion of the spin action in powers of $(U')^{-1}$. To lowest order we obtain an effective ferromagnetic interaction between the spins,

$$H_{\text{eff}} = -\frac{t^2}{U'}(\mathbf{n}_1 \cdot \mathbf{n}_2 + 1). \tag{9}$$

The third-order term in the expansion gives the interaction described by the real time Lagrangian,

$$L = \frac{t^2}{(U')^3}(\mathbf{n}_1 \cdot \mathbf{n}_2 + 1)\left[\int_{\mathbf{x}_1}^{\mathbf{x}_2} d\mathbf{x} \cdot (e\mathbf{E} - \mathbf{e}) + \delta H_{21}\right]^2. \tag{10}$$

The term $\propto \mathbf{E}^2$ is the spin contribution to the dielectric susceptibility, while the term linear in $\mathbf{E}$ describes the coupling of the external electric field to the spin-induced electric polarization:

$$L_E = \mathcal{E}\{(\dot{\mathbf{n}}_1 + \dot{\mathbf{n}}_2) \cdot [\mathbf{n}_1 \times \mathbf{n}_2] + 2(\mathbf{n}_1 \cdot \mathbf{n}_2 + 1)\delta H_{21}\}, \tag{11}$$

where $\mathcal{E} = \frac{t^2 e\mathbf{E}\cdot(\mathbf{x}_2 - \mathbf{x}_1)}{(U')^3}$. The second term in curly brackets describes the charge redistribution between the two sites

that takes place when $\mathbf{n}_1 \neq \mathbf{n}_2$, while the first term is the dynamical polarization originating from the coupling between the external and internal electric fields, $\mathbf{E} \cdot \mathbf{e}$, which for insulators was not considered before. Though weak, it leads to a number of unusual effects discussed below.

Physical consequences.—The dynamical part of L_E is eliminated by the rotation of the spins around $[\mathbf{n}_1 \times \mathbf{n}_2]$,

$$\begin{aligned} \delta\mathbf{n}_1 &= \frac{\mathcal{E}}{(S+1/2)}(\mathbf{n}_2 - (\mathbf{n}_1 \cdot \mathbf{n}_2)\mathbf{n}_1), \\ \delta\mathbf{n}_2 &= -\frac{\mathcal{E}}{(S+1/2)}(\mathbf{n}_1 - (\mathbf{n}_1 \cdot \mathbf{n}_2)\mathbf{n}_2), \end{aligned} \tag{12}$$

applied to the Berry-phase term in the spin Lagrangian,

$$L_B = \left(S + \frac{1}{2}\right) \sum_{i=1,2} (\cos\theta_i - 1)\dot{\varphi}_i, \tag{13}$$

where θ_i and φ_i are the polar angles describing $\mathbf{n}_i$. If the spin Hamiltonian H_S is not rotationally invariant, this transformation generates a coupling to electric field in the spin Hamiltonian. For example, the magnetic anisotropy,

$$H_S = \frac{A}{2}[(\tilde{S}_1^z)^2 + (\tilde{S}_2^z)^2], \tag{14}$$

where $\tilde{\mathbf{S}}_i = \mathbf{S}_i + \mathbf{s}_{i\alpha}$, gives rise to the interaction

$$H_{\text{int}} = \frac{A\mathcal{E}(t)}{\tilde{S}}[(\tilde{S}_1^z)^2 - (\tilde{S}_2^z)^2] \tag{15}$$

($\tilde{S} = S + \frac{1}{2}$). The time-dependent electric field induces transitions between the states with even and odd values of the total spin of the two sites S_t, while the projection of the total spin on the anisotropy axis S_t^z remains constant. These selection rules have to be compared with those for the conventional ESR where the time-dependent magnetic field leaves the total spin unchanged, while its projection on the static magnetic field changes by ± 1.

Consider now an arbitrary spin texture in a ferromagnet with $\mathbf{n}(\mathbf{x}, t)$ varying slowly at the lattice constant scale. The Lagrangian describing the linear coupling of the texture to the electric field is given by

$$\begin{aligned} L_E &= -\int d^3x T^{ab} E_a e_b(\mathbf{x}, t), \quad \text{where} \\ T^{ab} &= \frac{e}{(U')^3}\frac{1}{\upsilon}\sum_j |t_{j\beta,i\alpha}|^2 (x_j^a - x_i^a)(x_j^b - x_i^b) \end{aligned} \tag{16}$$

(υ is the unit cell volume). For a simple cubic lattice with the lattice constant a, $T^{ab} = \frac{g}{a^3}\delta^{ab}$ with $g = \frac{2ea^2t^2}{(U')^3}$. This interaction entirely comes from the dynamical part of Eq. (11), as the static polarization in the continuum limit is a total derivative.

Similarly to the two-spin case, the transformation $\mathbf{n}(\mathbf{x}, t) \to \mathbf{n}(\mathbf{x} + \mathbf{X}, t)$ with $\mathbf{X} = \frac{g}{\tilde{S}}\mathbf{E}$, applied to the Berry-phase term, cancels the interaction Eq. (16). Since this transformation leaves the Hamiltonian of a translationally invariant system unchanged, the effect of electric field is to shift a spin texture as a whole by the vector $-\mathbf{X}$. The shift is a small fraction of the lattice constant: $\frac{X}{a} \sim \frac{t^2 eEa}{(U')^3}$. It can, however, be enhanced by proximity to the metal-insulator transition (through larger t/U ratio) and by a larger distance between the spins a in magnetic semiconductors and quantum dot arrays. Although the coupling $T^{ab}E_a e_b$ is derived perturbatively assuming $t/U \ll 1$, it also holds in the regime of large charge fluctuations, since its form is constrained by crystal symmetries and invariance under global spin rotations, characteristic of nonrelativistic interactions.

A much stronger effect of this interaction is the resonant absorption of circularly polarized light by Skyrmions, magnetic bubbles, and magnetic vortices. These magnetic defects in two spatial dimensions carry a nonzero topological charge [18], $Q = \frac{1}{4\pi}\int d^2x \mathbf{n} \cdot \partial_x \mathbf{n} \times \partial_y \mathbf{n}$, integer for Skyrmions or bubbles and half-integer for vortices.

Magnetic vortices are spontaneously induced by magnetostatic interactions in nanodisks of ferromagnetic metals [19], while periodic arrays of magnetic bubbles appear in thin-film ferromagnets with a strong out-of-plane anisotropy upon application of magnetic field on the order of 10^2 Oe [20,21]. Similar arrays of Skyrmions, which are bubbles with "thick" domain walls, were recently observed in bulk ferromagnetic metals without inversion symmetry [22,23].

According to Eq. (16), a moving topological defect induces the net electric dipole moment in the direction normal to its velocity, $\mathbf{P} \propto gQ[\hat{\mathbf{z}} \times \dot{\mathbf{R}}]$, where $\mathbf{R} = (R_x, R_y)$ is the position of the center of the defect and $\hat{\mathbf{z}}$ is the unit vector normal to the film. Consider such a defect in a ferromagnetic insulator with a confined geometry, which breaks translational symmetry, e.g., a vortex in a nanodisk. We assume that the confining potential has the form $U = \frac{K}{2}(R_x^2 + R_y^2)$. In the adiabatic limit the dynamics of the collective coordinates R_i is described by Thiele equations [24–26]

$$G_{ij}\left(\dot{R}_j + \frac{g}{\tilde{S}}\dot{E}_j\right) + \alpha\Gamma_{ij}\dot{R}_j = -\frac{\partial U}{\partial R_i}, \tag{17}$$

to which we added the coupling of spins to the electric field. Here, α is the Gilbert damping constant and the nonzero components of the tensors G_{ij} and Γ_{ij} are

$$\begin{aligned} G_{xy} &= -G_{yx} = 4\pi Q \quad \text{and} \\ \Gamma_{xx} &= \Gamma_{yy} = \int d^2x \partial_i \mathbf{n} \cdot \partial_i \mathbf{n}. \end{aligned} \tag{18}$$

In the absence of electric field the (damped) eigenmode $\mathbf{R}(t) \propto (\cos\Omega t, -q\sin\Omega t)$ describes the rotational motion of the center of the spin texture with the frequency $\Omega = \frac{K}{4\pi|Q|}$ in the direction defined by $q = \text{sgn}(Q) = \pm 1$. The response to the rotating electric field

$$\mathbf{E}(t) = E_\omega(\cos\omega t, -\sigma\sin\omega t) \tag{19}$$

($\sigma = \pm 1$) at the resonant frequency, $\omega = \Omega$, is given by

$$X_\Omega = \frac{gE_\Omega}{2\tilde{S}} \begin{cases} \frac{i}{\Omega\tau} & \text{for } \sigma = +q \\ -\frac{1}{2-i\Omega\tau} & \text{for } \sigma = -q, \end{cases} \tag{20}$$

where $\tau = \frac{\alpha\Gamma_{xx}}{K}$ is the relaxation time. For $\Omega\tau \sim \frac{\alpha}{4\pi} \ll 1$ the excitation of the rotational motion by the electric field with $\sigma = +q$ is resonantly enhanced by the factor $\frac{1}{\Omega\tau}$ compared to the shift in translationally invariant systems, while for $\sigma = -q$ there is no enhancement. For magnetic insulators with $\alpha \sim 10^{-3}$–10^{-2} (see, e.g., Ref. [20]), the resonant enhancement by 3–4 orders of magnitude opens a possibility to manipulate spin textures with an electric field.

Discussion.—We now discuss why the hopping between filled and empty orbitals is essential to obtain the linear coupling of spins to electric field Eq. (11) and why such a coupling does not occur in the single-orbital model, in which only $t_{j\alpha,i\alpha} \neq 0$ and interactions between spins are antiferromagnetic. In that case

$$c_{21} = e^{\int_{\tau_i}^{\tau_f} d\tau [\langle \mathbf{n}_1(\tau)|\partial_\tau|\mathbf{n}_1(\tau)\rangle - \langle -\mathbf{n}_2(\tau)|\partial_\tau|-\mathbf{n}_2(\tau)\rangle]} \langle \mathbf{n}_1(\tau_f)| - \mathbf{n}_2(\tau_f)\rangle\langle -\mathbf{n}_2(\tau_i)|\mathbf{n}_1(\tau_i)\rangle. \tag{21}$$

A straightforward calculation shows that $c_{12} = c_{21}$. Since the $\mathbf{E}$-dependent term, C_{21}, is antisymmetric with respect to the permutation of the indices 1 and 2, there is no linear coupling of spins to electric field.

This result can be physically explained as follows. The first term in Eq. (11) originates from the spin dynamics in the virtual states with two electrons occupying the same site. More precisely, it describes the difference between the spin rotations in the state where both electrons occupy site 1 and the state with two electrons on site 2. In the single-orbital case, however, the virtual state of two electrons is a spin singlet independent of which site is doubly occupied and which is empty. Hence, no dynamical linear coupling to electric field. The hopping between partially filled and empty orbitals of transition metal ions is essential for the dynamic magnetoelectric coupling, which can be realized in a broad range of materials with either ferro- or antiferro-orbital orderings, such as the ferromagnetic Mott insulators $La_{7/8}Sr_{1/8}MnO_3$, $BiMnO_3$, La_2NiMnO_6, and La_2CoMnO_6 [27].

There are several interesting problems left for future studies. One is the relevance of the present mechanism for spin liquid states [28], where effects of the spin Berry phase are enhanced by strong spin fluctuations. Another issue of interest is the ring-exchange processes giving rise to persistent orbital currents in Mott insulators [29]. Dynamical effects resulting from the ring-exchange process deserve scrutiny.

In summary, we show that the dynamical spin Berry phase in multiorbital Mott insulators couples the electric field to the translational modes of spin textures. We derive equations of motion for the center-of-mass coordinates of Skyrmions and magnetic vortices in an applied electric field and predict the resonant absorption of the circularly polarized light by these topological objects as well as the ESR effect where spin transitions are induced by the time-dependent electric field.

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